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Page 1, lines 20 - 25 and page 2, lines 1 - 6.

Aι

The first scintillating crystal is calcium tungstate (CaWo<sub>4</sub>) which was used before the turn of this century to detect x-rays. The most significant discovery of a scintillating crystal is Thallium-activated sodium iodide NaI(Tl)) in the mid-40's. Even now, it is still the most widely used scintillating crystal. This is because large size crystals are readily available and quite inexpensive. Moreover, the light yield is the highest among all the known materials and is still the benchmark standard for all other scintillator crystals even after all these years. Even though NaI(Tl)) is widely used, it is not without problems. It is hygroscopic and very soft. Moreover, the density is too low (37 g/cm³), the effective mass number (Z<sub>eef</sub>) of 49 is also too small. It has a large and persistent after glow which interferes with the intensity counting system. Finally, the decay time about of 230 nanoseconds (ns) is too slow for many applications.

Page 2, lines 18 - 25 and page 3, line 1.

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BGO was found in the early 70's. It has higher density (7.13 g/cm<sup>3</sup>), and is non-hygroscopic. But it also has problems such as low light yield (15% that of NaI(Tl)), slow decay time (300 ns) and high refractive indices (n = 2.15) which results in light loss due to internal reflection. Still BGO scintillator crystals are now used in high energy calorimetry in particle physics research Institutes. It is also used as the detector for the 511 keV gamma-ray radiation of the positron emission tomographs (PET) in medical imaging application. Because of the low light and slow decay, the image produced from the BGO PET machine tends to be blurred with poor resolution.

Page 3, lines 2 - 10.

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In early 80's, the Ce doped GSO crystal was disclosed as a scintillator material. It has adequate density (6.71 g/cm<sup>3</sup>) and is also non-hydrogroscopic. The light yield is 20% of that obtained with NaI(Tl) with a much faster decay time (60 ns). Even though GSO crystals over 80 mm diameter have been produced, the crystal has not yet made in the PET marker because of a strong cleavage plane. It is very difficult to cut and polishing the crystal into any specific shape without the risk of fracturing of the entire crystal. Another unexpected problem is the high

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thermal neutron capture cross-section (49,000 barns) of the gadolinium. It will interfere with the gamma rays generated by neutron irradiation source. However, since there is no neutron involved in the PET process, gadolinium containing GSO is not a problem.

Page 3, lines 11 - 21.

P4

In the late 80's, the Ce doped LSO crystal was disclosed as a good scintillator material. Similar to GSO, it has high density (7.4g/cm³) and is non-hygroscopic. The light yield is significantly better and close to 75% that of NaI(Tl)) and the decay time is even faster (42 ns). The index of refraction is also very low (n = 1.82). Moreover, since LSO has a totally different crystal structure from GSO, it is fortuitous that in LSO structure, there is not any distinct cleavage plane making the material more suitable for detector block fabrication without the serious risk of fracturing. The thermal neutron capture cross-section is very low (84 barns) as compared to GSO. Lastly, it is now possible to commercially produce high quality, large size single crystals of LSO. Compared with all the other existing known scintillator crystals, Ce doped LSO seems to have the best combination of all the needed properties for PET or other high energy gamma-ray detector application.

Page 5, line 20 - 21.



Fig. 2 shows the LYSO scintillating light yield intensity as a function of lutetium concentration.

Page 5, line 22 - 23.

Rb

Fig. 4 shows the change of effective Z of LYSO as a function of lutetium concentration.

Page 5, line 25 and page 6, line 1.

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Fig. 2 is a pictoral representation of a typical cerium doped LYSO scintillating detector.

Page 6, lines 8 - 20.



In the background review, it is mentioned that Ce doped LSO has the best scintillating properties among all the known materials. But is still has a few serious problems to over come; namely, the isotope problem and the defect (deep trap) problem. In addition to these physical

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issues, LSO crystals also face two tough economic issues. First is the high melting temperature for growth. The melting point of LSO is estimated around 2200 °C . It is among the highest melting temperature crystals produced commercially. Special high temperature ceramics were used to build the furnace and Iridium crucibles were used to contain the melt for growth. The growth process is quite detrimental to both insulation and the crucible. High cost of frequent replacement of the hardware pushes cost too high to be bearable for practical use. Second is the high cost of raw material of lutetium oxide. It is not a common material. Moreover, the current material purity around 99.99% is not sufficient to guarantee consistent high light yield. It is highly desirable to reduce or even replace lutetium oxide as the main ingredient in new scintillator crystals.

Page 6, lines 21 - 25 and page 7, lines 1 - 4.

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The embodiment of this invention is to design a new crystal which can eliminate most of the problems of LSO crystal with sacrifice the scintillating properties. Our initial motivation is to reduce the growth temperature of LSO single crystals. It is a very difficult task to maintain the operation at such high temperature for long period of time (< 1 week). Since YSO has lower melting temperature near 2070 °C, we are seeking the possibility to find an intermediate composition (or LYSO composition) which may melt at lower temperature to ease the growth process. We also want to minimize the yttrium content to retain the LSO scintillating properties.

Page 8, line 4 - 6.

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A mixture of high purity fine powders consisting of 1476 grams of  $\underline{\text{Lu}_2\text{O}_3}$ , 359 grams of  $Y_2\text{O}_3$ , 317 grams of  $SiO_2$  and 3.6 grams of  $CeO_2$  were mechanically mixed thoroughly. The purity of  $Lu_2O_3$  is 99.99%. The purity of both  $Y_2O_3$  and  $SiO_2$  is 99.9999.

Page 8, line 19 - 25 and page 9, line 1.

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In order to check the melting behavior, an oversized crystal was grown in order to convert the maximum amount of melt to crystal. The resulting 30% LYSO crystal had a total length of 32 cm and a total weight of 1880.5 grams. The crystal took 8 days to complete the growth. In this case, the bottom 5 cm portion of the crystal contains a lot of visual inclusions and defects

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and thus was not usable for evaluation. It did, however, provide important information on the stability range of the melt.

Page 9, line 8 - 14.

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To evaluate the scintillating properties, two 10 mm slabs were cut from each crystals, one from the top of the crystal and one from the bottom right above the defect region. It has been found that the scintillating light yield of any LSO crystal decreases systematically form the top to the bottom of the boule. The relative light yield can vary by as much as a factor of 2. It appears that this is due to the impurities in the lutetium oxide (Lu<sub>2</sub>O<sub>3</sub>) source material. Since the LYSO also uses the same Lu<sub>2</sub>O<sub>3</sub> starting material, the LYSO will also show the same behavior

Page 9, line 15 - 21.

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The slab is placed under a Na<sup>22</sup> radiation source which generates the 511 keV gamma ray as the incident light. The scintillating light from the LYSO slab is captured by a Hamamatsu R877 photomultiplier. The scale used for the light output measurement is arbitrary unit. In this case, the light output for a standard NaI(Tl) scintillator is set a 100%. In comparison, the standard BGO crystal has a light output of 12.5%. The energy resolution is expressed as the full width at half maximum of the 511 keV gamma ray peak. The result of the measurement is summarized in Table 1 and plotted in Fig. 3.

Page 10, line 15 - 20.

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Fist, the top portion of each crystal has the best scintillating value n since the crystallization process has been found to be a purification process. The first (top) portion the result has many important implications of a crystal will have the least impurity content and thus the best performance. It is interesting to notice that the pure LSO crystal has produced light yield of 93% of that of NaI(Tl). This is significant higher than the published result of 75%. This value may approach to the ultimate scintillating power for LSO.

Page 11 line 9 - 13.

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In Fig. 3, it is shown that there is a linear reduction of light yield for the bottom portion of the LYSO and LSO crystals with linear increasing of lutetium content. This is the most direct

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evidence to show that the impurity is coming from the Lu<sub>2</sub>O<sub>3</sub> starting material. The phosphorescence is greatly reduced with more yttrium substitution and is nearly unnoticed beyond 50% substitution.

Page 12, line 3 - 11.

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Lastly, let us examine the issue of stopping power, radiation length and effective Z. For practicality, it is desirable to use material with highest Z and shortest radiation length.

Interesting enough, we find that in the case of LYSO, the effective Z increases rapidly with small substitution of lutetium. It will eventually slow down when it approaching to 100% substitution. Fig. 4 illustrates such effect. As a consequence, 30% LYSO has the same effective Z as NaI(Tl) and 60% LYSO has the same effective Z as GSO. In terms of the radiation length, we can save 30% lutetium with 10% increase of radiation length, save 50% and 70% lutetium with 1.5 and 2 times that of pure LSO. In other words, the reduction is not linear to the substitution. In fact, it is favor for the substitution.

Page 12, line 13 - 14.

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Fig. 2 illustrates the structure of the scintillation device with the crystal of the invention optically connected to the photomultiplier or other photon detector, which can include a photomultiplier tube, a PIN diode, and an APD(avalanche photo detector) diode.